1-19-2012

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Moti R. Paudel
Southern Illinois University Carbondale

Christopher S. Wolfe
Southern Illinois University Carbondale

Arjun K. Pathak
Southern Illinois University Carbondale

Igor Dubenko
Southern Illinois University Carbondale

Naushad Ali
Southern Illinois University Carbondale

See next page for additional authors

Published in *Journal of Applied Physics*, Vol. 111 No. 2 (2012) at doi: 10.1063/1.3676264

Authors
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JOURNAL OF APPLIED PHYSICS **111**, 023903 (2012)

Induced magnetic anisotropy and spin polarization in pulsed laser–deposited Co_2MnSb thin films

Moti R. Paudel,[1,a)] Christopher S. Wolfe,[1] Arjun K. Pathak,[1] Igor Dubenko,[1] Naushad Ali,[1] M. S. Osofsky,[2] Joseph C. Prestigiacomo,[3] and Shane Stadler[3,b)]
[1]*Southern Illinois University, Department of Physics, 483A Neckers, Carbondale, Illinois 62901, USA*
[2]*Naval Research Laboratory, Washington, DC 20375, USA*
[3]*Louisiana State University, Department of Physics and Astronomy, 202 Nicholson Hall, Baton Rouge, Louisiana 70802, USA*

(Received 12 August 2011; accepted 11 December 2011; published online 19 January 2012)

Co_2MnSb thin films were grown on glass and GaAs (001) substrates using pulsed laser deposition. The films were grown in magnetic fields ($H_G = 500$ Oe and 0 Oe) that were applied in the plane of the substrate during the deposition process. Angle-dependent magneto-optic Kerr effect measurements for films grown on glass revealed a uniaxial magnetic anisotropy in the direction of the applied growth field. Films grown on GaAs (001) exhibited more complicated magnetic anisotropy behavior, due to additional contributions from the substrate. Point contact Andreév reflection spectroscopy measurements indicated that the spin polarizations of the films were about $P \sim 50\%$, with negligible difference between films grown in zero and non-zero applied fields. © *2012 American Institute of Physics*. [doi:10.1063/1.3676264]

INTRODUCTION

The half-Heusler alloy NiMnSb was the first material predicted to be half-metallic.[1] Since this seminal discovery, many other materials have been predicted to be fully spin polarized, including many manganese-based full-Heusler alloys, such as Co_2MnSi and Co_2MnGe, and some quaternary full-Heusler systems, such as $Co_2MnSb_xSn_{1-x}$.[2–6] These systems have many attributes that make them attractive for potential spintronic applications, including large magnetic moments, high Curie temperatures, and a good lattice match with GaAs, a semiconductor. Unfortunately, even though the magnetic properties are appropriate for applications, the measured spin polarizations have been far below predicted values, in most cases, well below fifty percent. Assuming that the calculated electronic structures are correct, the reason for this shortfall has been attributed to many possible physical origins, one major cause being the occurrence of antisite disorder (i.e., the swapping of atoms from one sublattice into another).[10] In the case of the swapping of atoms from different magnetic sublattices, the magnetic moments of the misplaced atoms anti-align with the surrounding magnetic atoms, effectively reducing the total moment. Additionally, electronic structure calculations indicate that the formation of various antisites creates states in the minority gap, effectively destroying the half-metallic character of the ideal system, and significantly reducing the spin polarization.[10] The disorder in Co_2MnSi has been quantified experimentally using neutron and anomalous x ray scattering (Refs. 7–9) and studied theoretically through electronic structure calculations.[10] Since many of the techniques that are implemented to measure the spin polarization are surface sensitive (i.e., spin-resolved photoemission and point contact Andreév reflection), the reduced spin polarization in these systems could be attributed to a nonmagnetic surface region.

There are some strategies that can be implemented in order to reduce the antisite disorder or avoid its effects. Bulk crystal growth requires the alloys to be heated to their respective melting points and therefore introduces a minimum, thermally driven disorder. However, thin films can be grown at much lower temperatures (e.g., many Heusler alloy films are grown at $T \sim 180\,^{\circ}C$), and therefore the disorder may be reduced relative to that of melt-fabricated bulk materials. In addition, it is hypothesized that a strong magnetic field applied in a region local to the substrate during growth will affect the disorder.

Co_2MnSi films have previously been grown using a variety of techniques, including sputter deposition, pulsed laser deposition, and molecular beam epitaxy.[11,12] We have grown Co_2MnSb films on glass and GaAs (001) substrates using pulsed laser deposition (PLD). Magnetic fields (up to 500 Oe) were applied in-plane and localized at the substrate, so that the PLD plume dynamics would not be significantly affected. The magnetic properties of the films were then studied using angle-dependent (longitudinal) magneto-optic Kerr effect (MOKE) measurements. Coercive field (H_C) and remnant magnetization (M_R) was monitored as a function of angle to reveal the in-plane magnetic anisotropy properties of the films.

EXPERIMENTAL

Polycrystalline Co_2MnSb targets for pulsed laser deposition were fabricated using conventional arc melting methods (in an Ar atmosphere), using commercially available elements with 4 N purity. The targets (5 g buttons) were wrapped in tantalum foil and annealed at $850\,^{\circ}C$ for 48 h to enhance their homogeneity. The bulk targets were characterized structurally

a)Now located at the University of Alberta, Canada.
b)Author to whom correspondence should be addressed. Electronic mail: stadler@phys.lsu.edu.

using x ray diffraction (XRD) and were found to be in the full-Heusler $L2_1$ structure with negligible impurity phases. The magnetic, transport, and structural properties of bulk and thin film $Co_2MnSb_xSn_{1-x}$ were studied in detail elsewhere.[13–15] The targets were cut into thick disks and mirror polished using fine sand paper and ethyl alcohol. Prior to film deposition, the target surfaces were lightly ablated in order to remove any oxides or residues that remained on the surface when they were polished in atmosphere. The films were grown on fire-polished glass and on American flat GaAs (100) substrates. Before film growth, the substrates were first ramped to 600 °C (glass substrates only to 500 °C) and held at that temperature for 15 min in ultrahigh vacuum (UHV); In the case of GaAs, this procedure was intended to remove oxides and to reconstruct the surface. The growth temperature was 178 °C for all of the films.

The base pressure of the PLD growth chamber was less than 5×10^{-9} Torr. The targets were ablated with a Lambda Physik Compex 201 KrF excimer laser ($\lambda = 248$ nm). This laser has a pulse width of 20 ns and a 10 Hz maximum repetition rate. The target to substrate distance was 42 mm, and the fluence was approximately 4 J/cm^2. The deposition rates were estimated using a quartz crystal monitor, which was calibrated using Rutherford backscattering (RBS) measurements.

A UHV compatible electromagnet was fabricated and installed in the PLD chamber, so that films could be grown in applied magnetic fields. The magnet could sustain a static field up to H = 650 Oe. An electromagnet with an iron-silicon alloy core was employed so that the field could be localized at the substrate, i.e., so that the PLD plume would not be significantly affected by stray fields. The magnetic poles had a gap of about 2 cm, and the substrates were attached to a radiant-conduction copper stem heater that was inserted between the magnet poles. For the films grown in this study, the static applied field was H = 500 Oe.

Angle-dependent (in-plane) magnetic hysteresis measurements were made at room temperature using a magneto-optic Kerr effect (MOKE) system that utilized a longitudinal geometry and employed a HeNe laser. The measurements were taken every five degrees, and the loops were normalized to the saturation values. The remnant magnetizations (M_R) and coercive fields (H_C) were extracted from the data and plotted as a function of angle.

RESULTS

Angle-dependent, longitudinal MOKE measurements made on a 500 Å Co_2MnSb film grown on a fire-polished glass substrate are shown in Fig. 1. The film was grown in a localized, in-plane magnetic field of $H_G = 500$ Oe (see the schematic of the PLD growth geometry in Inset A of Fig. 1). The MOKE measurement angle (θ) in Fig. 1 was measured relative to the direction of the applied field (Fig. 1, Inset B). The most square hysteresis loop (the squareness is defined as M_R/Ms, where M_R is the remnant magnetization and M_S is the saturation value) was measured in the direction of the applied growth field. We estimated the angular error (offset) in our measurement to be $\Delta\theta \approx \pm 7.5°$, which includes the combined error in the growth field angle and the MOKE measurement angle.

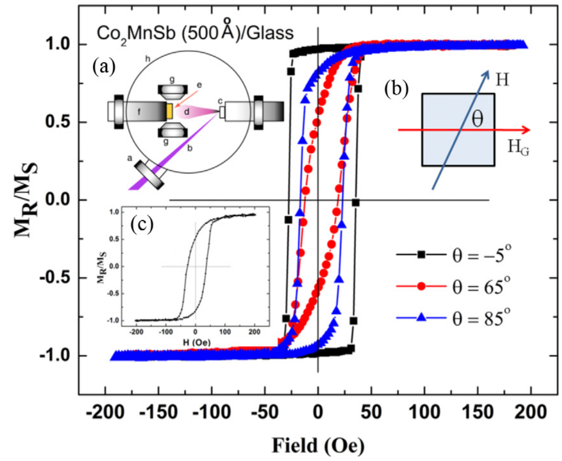

FIG. 1. (Color online) Room temperature, longitudinal MOKE measurements (M/M_S) of Co_2MnSb(500 Å)/glass. (Inset A) A schematic of the PLD system with magnet: (a) UV laser window, (b) focused UV laser beam, (c) target, (d) plasma plume, (e) heated substrate, (f) heater assembly, (g) magnet pole face or coil (x2), and (h) UHV vacuum chamber. (Inset B) Sample and field geometry: The angle θ is the angle between the direction of the growth field ($H_G = 500$ Oe) and that of the in-plane applied MOKE field (H). (Inset C) Representative hysteresis loop for Co_2MnSb(500 Å)/glass grown in zero field.

Angle-dependent MOKE was measured full circle in increments of 5°. M_R/M_S and H_C were calculated and plotted as a function of θ (see Fig. 2). Both M_R/M_S and H_C had similar angular dependencies, with maximum values in directions that were nearly collinear with the growth field (within ±7.5°). The largest coercive field was $H_C = 30$ Oe, and the minimum value of $H_C < 10$ Oe was measured at $\theta \approx 65°$. Another feature was observed (in both M_R/M_S and H_C) perpendicular to the growth field direction ($\theta \sim 85°$), indicating a four-fold contribution to the anisotropy. The angular dependence differs from that observed in similar studies, where the perpendicular maximum is absent.[15,16] It should be noted that the films were cycled through numerous complete hysteresis loops (i.e., each point in Fig. 2 represents the average of at least 10 saturating field cycles), meaning that the anisotropy is a (semi-) permanent feature of the film. Films grown on glass in the absence of an applied field (with all other growth and measurement parameters being identical) had a comparatively weak in-plane angular dependence, attributed to either shape anisotropy or slightly asymmetric deposition geometry. Similar results were obtained for a 1000 Å film grown with $H_G = 650$ Oe and for slowly oscillating fields. The field values were otherwise limited to 500 Oe, due to heating and outgassing for higher values.

The strong uniaxial component of the anisotropy could be attributed to texturing during the growth process, such as that observed in sputter-eroded iron films.[17] However, in the case of Co_2MnSi films grown epitaxially on Si(001), a uniaxial contribution to the anisotropy was only observed for films grown at grazing-incidence fluxes.[18] Films grown at normal incidence did not exhibit uniaxial anisotropy. In this study, all Co_2MnSb films were grown with the PLD plume at normal incidence with respect to the incident plume direction, and the substrates were positioned in the center of the plume.

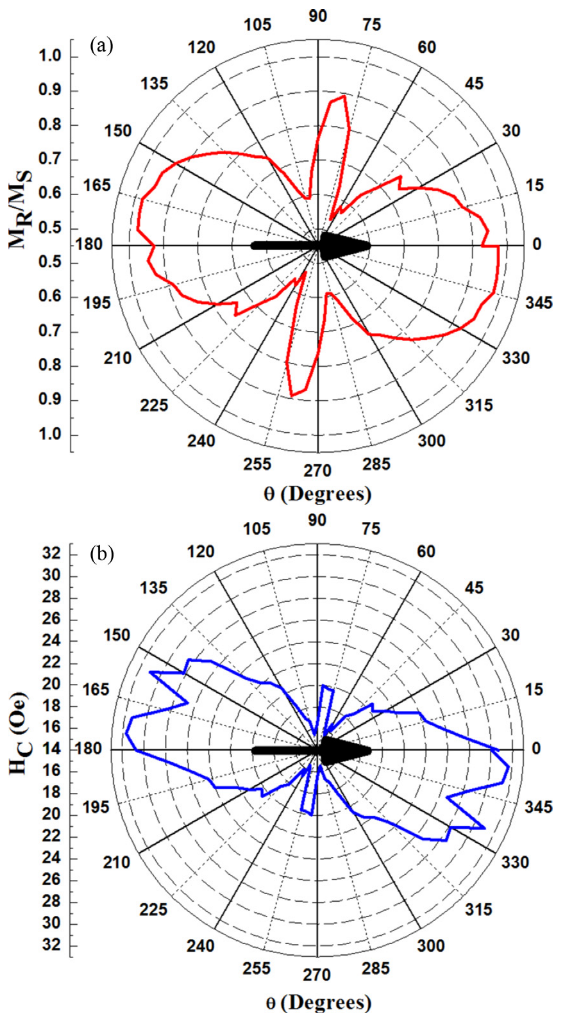

FIG. 2. (Color online) The angular dependence of (a) M_R/M_S and (b) H_C for Co_2MnSb(500 Å)/glass calculated from room temperature MOKE data. The black arrows at the centers indicate the direction of the growth field (H_G).

XRD measurements did not show a significant difference between field-grown and zero-field-grown films.

Room temperature MOKE loops for a 500 Å Co_2MnSb film grown on a GaAs (001) substrate are shown as a function of angle in Fig. 3. For the film grown in field, $H_G = 500$ Oe and was directed along the [01-1] direction (easy axis). The film grown in zero field exhibited magnetic anisotropy affects that are consistent with previous studies of this material.[15] Films grown in field showed stark differences. The MOKE loops along the easy axis [01-1] for films grown in zero field and in a field $H_G = 500$ Oe are shown in Fig. 3(a). Both loops are very square; however, the coercive field has been reduced from about $H_C = 50$ Oe for zero field to about 30 Oe for field-grown. Similar reductions (but to a lesser degree) were observed for the [010] and [001] directions (Figs. 3(b) and 3(d), respectively). The largest difference occurs for the MOKE loops measured along the [011] or hard-easy axis (Fig. 3(c)). In this case, the coercive field of the field-grown sample is less than 10 Oe and is slightly shifted along the field axis, an effective exchange bias, the origin of which is not known. Therefore, in this case, the in-plane hard axis becomes that which is oriented perpendicular to the growth field, H_G.

One objective of this study was to determine whether growth in applied magnetic fields affects the degree of spin polarization of materials. It was hypothesized that the applied field would enhance the magnetic order (and possibly the crystalline order), since the PLD growth process occurs in non-equilibrium conditions. We have measured the spin-polarizations of the films grown in field ($H_G = 500$ Oe) and zero field, using the point contact Andreév reflection technique.[19] The spin-polarization was estimated to be slightly higher than 50%, with no significant difference between the films grown in field versus those grown in zero field. Experiments employing planar tunneling techniques to measure the spin polarization are currently underway.[20]

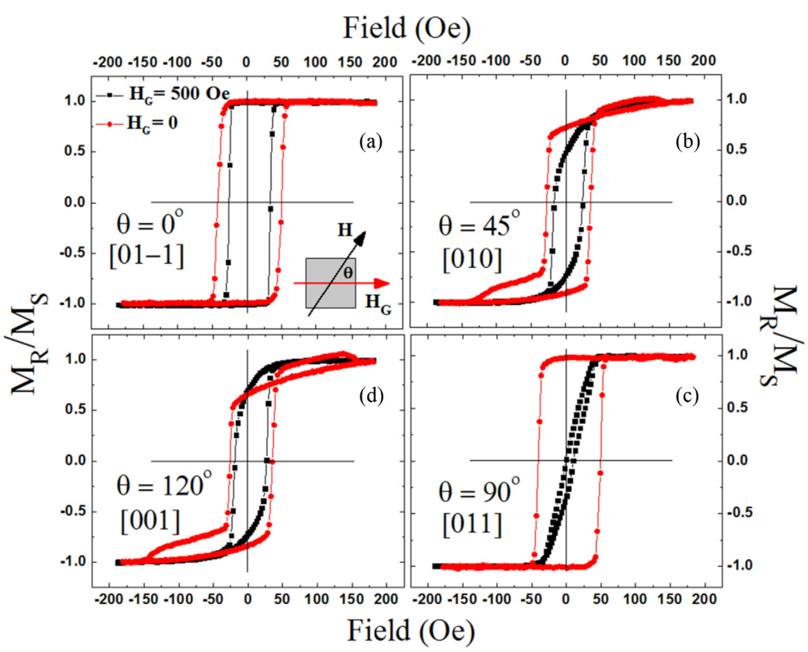

FIG. 3. (Color online) The angular-dependent MOKE loops of a 500 Å Co_2MnSb film grown on GaAs (001). The growth field was $H_G = 500$ Oe and was directed along the direction of the easy axis [01-1]. The angle θ is the angle between H_G and the applied MOKE field (H) as defined in the inset of A.

In conclusion, Co_2MnSb thin films were grown on fire-polished glass and GaAs (001) substrates in zero field and in applied fields up to $H_G = 500$ Oe. An induced uniaxial and four-fold anisotropy was observed in the films grown in field on glass with the easy axis parallel to the growth field direction. Films grown on GaAs (001) (in the [01-1] direction) were also strongly affected by the growth field; the easy axis was found to be parallel to the growth field and the hard axis perpendicular to it. The spin polarizations of films grown on GaAs were determined, using Andreév reflectivity, to be about 50%, with no significant difference between films grown in field and those grown in zero field. More information might be acquired from planar tunneling techniques, and studies of the growth of films in much higher magnetic fields are underway.

ACKNOWLEDGMENTS

This work was supported by the National Science Foundation (Grant No. NSF-DMR-0545728).

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